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# REMOVAL OF URANIUM AND RADIUM FROM SOLUTION WITH BACTERIALLY-PRODUCED IRON SULPHIDE PARTICLES USING HIGH GRADIENT MAGNETIC SEPARATION

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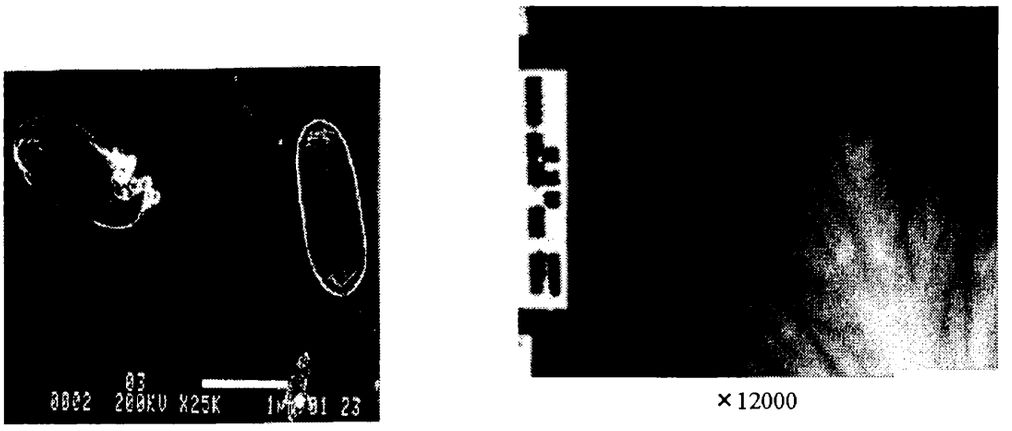
Today, water pollution by heavy metals is one of the challenges for us. Recently, J. H. P. Watson found that iron sulphide FeS nano-particle produced by sulphate reducing bacteria is an excellent adsorbent for a wide range of metal ions in solution [1]. In order to evaluate the applicability for the FeS adsorbent to the radioactive ions in solution, in this article, we studied adsorption characteristics to the uranium U and radium Ra ions. This bacteria form FeS layer having magnetic and non-magnetic component on their surface. The magnetic component of the FeS were collected with an open gradient magnetic separator OGMS and adsorbed U and Ra ions in well-water at Ningyou-toge uranium mine to the magnetic FeS adsorbent were removed by the technology of the high gradient magnetic separation HGMS.

A kind of sulphate-reducing bacteria *Desulfovibrio Vulgaris Miyazaki* as shown in Fig. 1 were cultured with a medium modified by addition of iron sulfate in an anaerobic incubator. The temperature was kept at 310 K. They form FeS layer having excellent adsorption characteristics for heavy metal ions in solution around their surface. The electron microscope micrographs were taken with a JEM-2000FX operating at 200 kV accelerating voltage.

To select the bacteria which produce a magnetic FeS as the adsorbent for the high gradient magnetic separation, an open gradient magnetic separator OGMS with a 10 Tesla superconducting magnet was used [2]. Collected bacterially-produced magnetic FeS particles were wet, therefore, they were dried by several methods to use as an adsorbent, i.e. dried at room temperature, by heat treatment and by freeze dry, in an anaerobic condition respectively. For magnetization measurements for these magnetic FeS particles, a SQUID magnetometer MPMS – 7 was used.

To evaluate adsorption properties of the magnetic FeS particles for U and Ra ions, sample water having 6 ppb U and 5-9 x $10^{-4}$ Bq Ra concentration was extracted from an well as shown in at Ningyo-Toge mine in Okayama Prefecture. U ion concentration was evaluated by an ICP-MS spectrometry and Ra ion concentration was evaluated by radioactivity measurements. To the water, 100 ppm of magnetic FeS particles were used for adsorption.




a) Sulphate reducing bacteria *Desulfovibrio Vulgaris Miyazaki.*

b) Magnified micrograph of a FeS layer surface.

**FIGURE 1. Transmission electron microscope micrograph of bacteria and typical bacterially-produced FeS layer on the surface.**

After 5 hour stirring, these sample waters were introduced to a superconducting HGMS having a 10 Tesla superconducting magnet as shown in Fig. 2. The HGMS has SUS 430 magnetic stainless steel filters. Strand diameter of the filter is 100 μm.

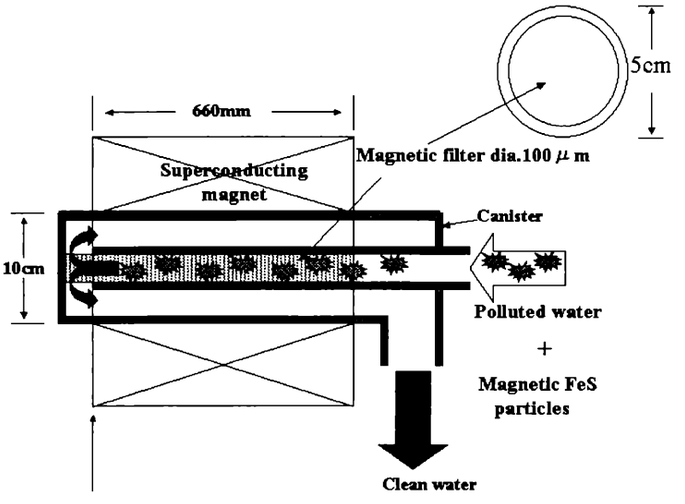



FIGURE 2. High gradient magnetic separator HGMS to remove heavy metal ions from polluted water. U ions are adsorbed on the surface of magnetic iron sulphide FeS layer. Magnetic 430 stainless steel filter strand diameter is 100 μm.

A transmission electron microscope micrograph of a typical bacterially-produced FeS layer is shown in Fig. 1 b). A magnified micrograph indicates that the surface area of the layer is

extremely large.

Magnetization curves obtained for dried bacterially-produced magnetic FeS particles were shown in Fig. 3 with data collected at 2 Tesla field strength. The influence of the drying conditions upon the magnetization is small. Magnetic susceptibility of the particles collected at 2 Tesla is larger than the particles collected at 10 Tesla. Therefore, strong magnetic FeS particles can be collected by the low field collection. In this experiment, the particles collected at 10 Tesla were used.

Experimental results of U ion cleaning with the high gradient magnetic separator HGMS shown in Fig. 2 at 10 Tesla are listed in Table 1 evaluated with the ICP-MS spectrometry. The U ions in the well water are reduced from 6.3 ppb to 0.66 ppb by the 100 ppm adsorbent addition, which corresponds to the reduction rate of 90 %. Thus it was revealed that these magnetic FeS particles are good adsorbent for U ions. Adsorption characteristics for the isotope ions of U, i.e., U-234, U-235 and U-238 ions, analyzed by radioactivity measurements are summarized in Table 2. U-234 and U-238 ions are removed to around 10 % of the initial value by the 100 ppm adsorbent addition followed by the 5 hour stirring and the HGMS at 10 Tesla.

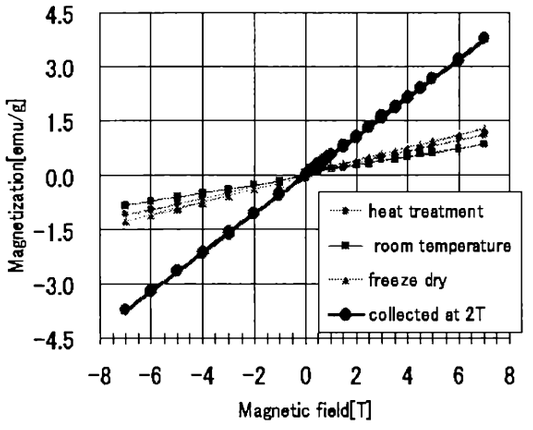



**FIGURE 3. Magnetization curves obtained for dried bacterially-produced magnetic FeS particles collected at 10 T with the data collected at 2T .**

**Table 1 Uranium concentration evaluated by an ICP-MS spectrometry before and after the experiment.**

| Initial Concentration | After HGMS |
|---|---|
| U ( $\mu$ g/L) | U ( $\mu$ g/L) |
| 6.3 | 0.66 |

To conclude, with bacterially-produced magnetic iron sulphide adsorbent followed by the high gradient magnetic separation at 10 Tesla, uranium ions in well water at an uranium mine could be reduced from 6.3 ppb to 0.66 ppb with using 100 ppm of the adsorbent.

Radioactivity analysis results also indicated that similar reduction rate were obtained for uranium isotope U-234 and U-238 ions to 10 % from the initial concentration value.

Experimental results for radium removal will be shown in the conference.

ACKNOWLEDGEMENTS

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REFERENCES

[1] J. H. P. Watson et al., "Structural and magnetic studies on heavy-metal-adsorbing iron sulphide nanoparticles produced by sulphate-reducing bacteria", J. Magnetism and Magnetic Materials, 214, 13-30 (2000).

[2] D. Ito et al., "Removal of As, Cd, Hg and Pb Ions from Solution by Adsorption with Bacterially-produced Magnetic Iron Sulphide Particles", IEEE Trans. on Applied Superconductivity, vol.14, p.1551 (2004).

**Table 2. Radioactivity analysis of uranium isotopes for different sampling site..**

| | Nuclide | Adsorbent (ppm) | Time (h) | Initial activity x $10^{-5}$ Bq/cm$^3$ | After HGMS x $10^{-5}$ Bq/cm$^3$ |
|---|---|---|---|---|---|
| No.1 | U-238 | 100 | 5 | <1.2 | <0.6 |
| | U-235 | 100 | 5 | <1.2 | <0.6 |
| | U-234 | 100 | 5 | 1.3 | <0.6 |
| No.2 | U-238 | 100 | 5 | 4.1 | <0.6 |
| | U-235 | 100 | 5 | <1.2 | <0.6 |
| | U-234 | 100 | 5 | 5.7 | <0.6 |
| No.3 | U-238 | 100 | 5 | <1.2 | <0.6 |
| | U-235 | 100 | 5 | <1.2 | <0.6 |
| | U-234 | 100 | 5 | <1.2 | <0.6 |